Atmospheric Science Program

Program Description

May 3, 2004

The Atmospheric Science Program (ASP) has both a long-term goal, and a specific science focus that changes from time to time according to national and DOE needs. The long-term goal is to develop a comprehensive understanding of the atmospheric processes that control the transport, transformation, and fate of energy related chemicals and particulate matter, especially in the context of climate change. Beginning in FY 2005 the specific science focus will be **aerosol radiative forcing of climate. Associated with this focus is the objective of enhancing the scientific knowledge needed to simulate and predict radiative forcing and other climatic effects of aerosols.**

Projects in atmospheric chemistry and environmental meteorology hitherto supported under the previous program focus on air quality will no longer be supported, except in the context of this new science focus.

It is expected that a new grant solicitation notice will be published very soon, with proposals due on or about June 21, 2004, for funding beginning in November, 2004.

Stephen E. Schwartz of Brookhaven National Laboratory has been named the ASP Chief Scientist. A Science Steering Committee (SSC) will be formed once the Science Team has been selected. The Science Team will consist of principal investigators and key associates for all ASP-funded projects, as well as non-ASP funded participants selected from other programs and other agencies.

Much of the following description of the ASP is based in part on a report by a panel of DOE's Biological and Environmental Research Advisory Committee (BERAC). This report is expected to be published soon and be made available on the DOE Office of Science website http://www.science.doe.gov/. However, this Program Description does not include at this time all of the areas of research dealing with aerosol radiative forcing that were recommended by the BERAC panel.

I. INTRODUCTION

The DOE Atmospheric Science Program (ASP) is a component of the U.S. Climate Change Science Program (CCSP). The objective of ASP is to provide and improve the scientific knowledge needed to simulate and predict radiative forcing by aerosols and their effects on climate. The role of aerosols in climate forcing is a critical factor in climate change assessment, as well as an essential element in advancing the state of the art in climate modeling. Aerosol forcing appears to be the same order of magnitude as the effect of greenhouse gases, but far more uncertain. The forcing has two major components, direct and indirect.

Direct effects of aerosols are the influence of the aerosols on the Earth's radiation balance due to the scattering and absorption of radiation by particles in clear (cloud-free) air. Indirect effects of aerosols include their influence on the radiation balance and hydrology through their impact on cloud microphysical properties (first indirect effect) and amount (second indirect effect). There is also a semi-direct effect, in which the heating by aerosol particles due to absorption of solar radiation decrease cloud amount.

The direct aerosol forcing is much better understood and quantified than the indirect forcing, especially forcing due to sulfate aerosols. Aerosol indirect effects have been demonstrated and quantified in numerous instances, but the indirect forcing is much more uncertain than the direct forcing.

ASP will focus on two areas of large uncertainty: (1) uncertainties associated with the loading, distribution, and fate of atmospheric aerosols, and their chemical and microphysical properties that affect the absorption and scattering of radiation, and (2) uncertainties associated with direct and indirect effects of aerosols on radiation and cloud properties. ASP will include examination of the amount, distribution, chemical and optical properties of carbonaceous aerosols (organic and elemental) in addition to inorganic aerosols (mainly sulfates and nitrates) associated with energy-related activities.

Both of these areas involve complex questions of atmospheric chemistry and physics as well as transport and transformation issues that need to be investigated in depth to improve the understanding of aerosol climate forcing and reduce uncertainties.

<u>ASP will support only research that can be directly related to aerosol radiative forcing of climate</u>.

ASP will consist of four major elements. These key elements are: (1) focused laboratory experiments, (2), field measurements, (3) fundamental theoretical and process modeling, and (4) the development of new instruments and methods to better measure the composition, optical, and cloud nucleating properties of atmospheric aerosols, in support of elements 1 and 2.

ASP will work closely with two other DOE climate change programs, namely the Atmospheric Radiation Measurement (ARM) Program and the Climate Change Prediction Program (CCPP) and will be closely coordinated with other relevant federal agency programs, both within and beyond the CCSP.

II. ELEMENTS OF THE PROGRAM

Within each of the four elements of the program, relative priorities have been identified and enumerated.

LABORATORY STUDIES

Laboratory studies are required to provide a basis of understanding of the processes governing aerosol formation and transformation in the atmosphere affecting their optical and cloud nucleating properties. This understanding is required to represent these processes in chemical transport models. ASP will support laboratory studies directed to these issues. Laboratory studies must demonstrate relevance to what actually goes on in the atmosphere, and they should provide explicit guidance as to how these processes and their importance to aerosol radiative forcing can be verified via field studies.

To elucidate both the indirect effects of aerosols on climate as well as the contribution of carbonaceous species to radiative forcing, directed and focused laboratory experiments are needed. The optical properties of aerosols as well as their ability to function as cloud condensation depend on their chemical composition as well as size. Chemical composition includes the distribution of chemical species within the aerosol particle itself because, for example, the presence of hydrophilic materials on the aerosol surface might promote water uptake while the presence of surface hydrophobic materials might decrease it. Such interfacial structure might not only alter water uptake, but also the kinetics and mechanisms of chemical changes that lead to the conversion of a hydrophobic to a hydrophilic surface and thereby affect their role as potential cloud condensation nuclei (CCN).

Organic materials from biogenic sources may be important as significant sources of carbonaceous aerosol, and the processes governing their contribution to the atmospheric aerosol burden must be determined.

In the ambient atmosphere the chemical composition and the physical and optical properties of particles will change with time because of various chemical and physical aging processes. These changes can dramatically alter the magnitude of the direct and indirect effects of a given aerosol loading, and these governing processes must be understood.

It is essential that laboratory experiments be closely coupled to field measurements to investigate the physical and chemical evolution of particles and how this evolution alters their radiative and cloud-nucleating properties. This will allow interpretation of the field data and appropriate translation into models that will accurately reflect the processing of particles in the atmosphere and provide the needed capabilities to simulate and predict how much these changes affect the direct and indirect radiative forcing by aerosols.

Following are examples of the types of laboratory studies that will be useful in coupling the results of field studies to models and in providing and improving understanding of key processes that are or need to be represented in models. The order provides a rough guide to the relative priorities of the kinds of laboratory studies that would be funded by the ASP.

1. There are large uncertainties in the role of secondary organic aerosols and carbonaceous aerosols as CCN, because atmospheric processing can change their physical properties. These changes may also have an effect on their life cycle in the atmosphere. There is therefore a need for measurement of kinetics, mechanisms, and products of the oxidation of organic aerosol constituents and their precursors by tropospheric oxidants (e.g., OH, O_3, NO_3) for different forms of organics in the absence and presence of inorganics (e.g., aqueous NO_3 and SO_4). Included in this area would be identification of interactions between natural and anthropogenic aerosols and precursors (e.g., acid-catalyzed reactions of organics in particles) and elucidation of the role of trace gases such as NO_x in the formation and further oxidation of organic aerosols.
2. Research to improve understanding of interactions of organic and organic-coated aerosols with water vapor and liquid water, as well as the uptake and reactions of trace gases such as HNO_3, SO_2, etc., that are known to modify the optical properties of an aerosol particles as well as their ability to function as a CCN.
3. Research to understand the potential of organics and mixed particles to serve as CCN and ice-forming nuclei (IN), including the effects of changes in their properties induced by chemical processing or “aging” of the particle.
4. Determination of optical properties of particles of mixed composition and phase including both inorganics as well as organics (e.g., non-spherical particles of sulfate and water layers on soot, liquid layers surrounding solid cores, surfactant layers on aqueous and solid substrates).
5. Research to understand nucleation mechanisms and how to represent them in models, including mechanisms of new particle formation and nucleation involving trace gases such as ammonia (NH_3) and volatile organic compounds.
6. Identification of "markers" for natural vs. anthropogenic components of ambient aerosols. These are important because source attribution is a key to understanding the relative importance of various sources of atmospheric aerosols.
7. Research to better understand the chemical characteristics of particles designated as “black carbon” by current methods. This is important because “black carbon” is chemically complex and is defined in a number of ways.

FIELD STUDIES

Field studies are an essential component of any comprehensive aerosol research program. Real world measurements are the only means to identify and quantify processes that occur in the actual atmosphere, evaluate performance of models, and evaluate remotely-sensed data retrievals. It is anticipated that many if not most of the ASP field studies will be done in collaboration with aerosol research programs in other agencies.

The design of field measurements addressing aerosol forcing issues may require different approaches ranging from basic sustained monitoring through highly intensive, sophisticated short-term campaigns. Three classes of field programs are needed: (1) long-term systematic measurements linked with ARM sites or their equivalent, supplemented by satellite data as well as periodic, regularly scheduled aircraft observations; (2) systematic ground based measurements supplemented by periodic intensive studies aimed at characterizing long range transport, mega-city plumes, or aerosol life cycles and; (3) highly intensive atmospheric, process-oriented campaigns to test specific hypotheses about aerosol-radiative interactions, especially indirect effects involving cloud processes.

It is unlikely that ASP will have the resources to conduct long-term systematic measurements. Therefore, we expect to leverage long-term measurements conducted by other programs, e.g., ARM, and work collaboratively with other programs, while focusing on more intensive studies and process-oriented campaigns.

It is anticipated that conducting field studies will be a principal focus for the DOE laboratories, with participation by other science team members wherever appropriate. Once the ASP Science Team has been formed, proposals for specific field studies will be solicited that are coordinated with individual laboratory and modeling studies, to make the best use of the relevant capabilities from the ASP Science Team, and in collaboration with scientists funded by other federal agencies and the private sector. These field study proposals will be reviewed and a Lead Scientist for a given field study will be designated and given responsibility for leading the implementation of the study.

Following is a rough guide to the kinds of field studies that would be funded by the ASP. Indeed, the ASP is expected to participate in a mix of ASP-led and other-agency-led field studies, e.g., the upcoming Mexico City Study. It is also expected that studies will be conducted in conjunction with ARM sites, and especially in conjunction with the ARM Mobile Facility.

The following indicates the scientific scope and priorities of field studies of greatest interest to DOE at this time.

- <u>Life Cycle of Aerosols</u>. Characterization of the formation of aerosols, and the evolution of their chemical, microphysical, and optical properties as they are transported downwind from different types of aerosol and aerosol precursor sources, such as urban centers, forest fires, and power plants. It is important that these studies be conducted in different environments so that the full range of processing conditions is understood. It is important to understand the evolution of aerosol properties over a period of time. Coupled with such process-focused studies should be measurements of the vertical profiles of aerosol optical properties such that the radiative forcing of aerosols can be determined. Investigation of the indirect effect of aerosols on clouds in plumes downwind of isolated sources or cities to understand the aerosol effects on precipitation (e.g., rainfall suppression) and precipitation mechanisms would also be desirable in these studies. Also needed are investigations to determine the importance of chemical processing to removal and possible transformation from hydrophobic to hydrophilic particles.

- <u>Indirect Effects</u>. Investigation of the effects of aerosols on clouds of various types, to develop a better understanding of both the first and second indirect effects. Such investigation should include the relationship between aerosol composition/microphysics and CCN concentrations, CCN concentrations, updraft velocities, and cloud droplet concentrations, and the relation between cloud droplet microphysics and the presence of drizzle. There is also a need for research on the effects of entrainment and other mixing processes on cloud microphysics as well as their importance in aerosol removal.

In addition to addressing these priorities, it is expected that ASP will participate in selected radiation budget closure experiments, e.g., in conjunction with ARM and/or in conjunction with megacity studies, to test our ability to predict the direct effects of aerosols given information on their size distribution, vertical concentration profiles, and chemical characteristics. ASP may also support research on meteorological processes important to the transport, removal, and vertical distribution of aerosols.

MODELING

The climate modeling community need improved process models of aerosols, for both direct and indirect effects, to better account for the effects of aerosols on regional and global climate.

The need is for process models applicable to scales from local to global. These models need to incorporate the chemistry and microphysics that governs the formation, radiative properties, cloud-nucleating properties, and fate of carbonaceous, inorganic, and mixed carbonaceous/inorganic aerosols. It is especially important that knowledge about the mechanisms controlling the lifetime and removal rates of aerosols be represented in models so as to better simulate quantitatively the concentration and distribution and radiative and cloud-modifying properties of atmospheric aerosols.

There are two areas in which additional modeling is particularly needed.

- Regional scale models are needed to simulate the evolution of particle size distribution and composition with integrated meteorological processes so that field program results can be used effectively to evaluate model-based representation of the pertinent aerosol chemical and microphysical processes. These models must include cloud and precipitation processing of aerosols as well as clear-air processes. These models should include both natural and anthropogenic aerosols and aerosol precursors. Process models that allow comprehensive testing of aerosol evolution in the atmosphere and quantitative comparisons with field or laboratory data are also required.

- Cloud-resolving models and higher-resolution models such as large-eddy simulation models with detailed microphysics for coupled cloud/aerosol systems are needed to investigate indirect effects of aerosols on clouds. While ASP is not expected to develop cloud-resolving or large-eddy simulation models, ASP scientists are encouraged to collaborate with scientists who have developed or are developing such models, in order to improve and test those models.

It is expected that all of these modeling efforts will be coordinated with the CCPP in order to effectively incorporate the anticipated modeling improvements into GCMs. Modeling studies should have explicit ties to design and interpretation of ASP field studies, rather than be stand-alone projects. Emphasis will be on improving models that also include chemistry and more realistic treatment of cloud microphysics.

INSTRUMENT DEVELOPMENT

This program element necessarily has a fairly broad scope. It is anticipated that the most promising new instrument ideas will be funded according to these relative priorities. Opportunities for additional funding are available through DOE's SBIR Program.

Better assessments of the indirect effect of aerosols on clouds and of the direct effects of aerosols on the climate require several instrument developments or refinements. Each of these instrument development needs is assigned a priority. Priority 1 is the highest category, and priority 3 is the lowest. Instrument development projects will be expected to participate in and contribute to ASP field studies as new instruments and capabilities are tested.

In general, there is a **priority 1** need for the development of lightweight compact instruments suitable for deployment on research aircraft used in field studies.

There are several key aerosol properties or quantities related to aerosol formation and growth, which cannot be currently measured or for which current instrumentation is inadequate.

- **Size-Resolved Chemical Composition**. There is a need to develop improved measurement methods to characterize the bulk and the size-resolved chemical composition of ambient aerosols in real time, particularly for carbonaceous aerosols. Improved measurements would facilitate the identification of the origin of aerosols, *i.e.* primary versus secondary and fossil fuel versus biogenic. Such measurements would help to elucidate how aerosol particles are processed in the atmosphere by chemical reactions and by clouds and how their hygroscopic properties change as they age. This is a **priority 1** instrument need since relatively little is known about organic and absorbing particles that are known to be abundant in many locations in the atmosphere. There is a need for instruments suitable for real-time measurements of the composition of particles at the molecular level. There have been important recent advances in the development of such instruments that include particle mass spectrometers and single particle analyzers. These new instruments still have important limitations. Some of those limitations are in the quantification of black carbon vs. organic carbon, in the speciation of refractory and volatile organic compounds, and in the calibration of both organic and inorganic components. Important remaining needs are: (1) quantifiable results over a wide range of compounds, a problem for laser ablation aerosol mass spectrometer methods; (2) measurements over a range of volatility so that dust, carbon, and salt are detectable, a problem for thermal decomposition aerosol mass spectrometers; (3) speciation of individual organics, including those containing oxygen, nitrogen, and sulfur; (4) identification of elemental carbon and other carbonaceous material so the makeup of the absorbing fraction is known; (5) measurements with high time resolution, an inherent problem with filter techniques; (6) identification of source markers such as isotopic abundances in aerosols and; (7) the ability to probe the chemical composition of aerosol surfaces.

- **Aerosol precursors**. There also are several improvements in gas phase chemistry that are needed to further understanding of aerosols and clouds. Gas phase measurements for H_2SO_4, a major aerosol precursor, have revealed a wealth of new information in the last decade. To make further progress, fast measurements are needed of NH_3, ion clusters, and gas phase organics that might either condense or dissolve into preexisting aerosols or cloud droplets. Development of instruments that will allow such measurements are **priority 1** since they are critical in advancing understanding of aerosol evolution.

- **Aerosol absorption**. The aerosol absorption coefficient, together with the aerosol scattering coefficient, determine the single scattering albedo. This key aerosol property and the factors that contribute to it are critical to determine heating rates and climate forcing by aerosols. Development of reliable instruments for the *in situ* measurement of the single-scattering albedo covering the solar wavelengths (UV, visible, and near infrared) is needed for particles containing black and organic carbon, dust, and minerals. Such measurements must be made *in situ* without altering aerosol properties, including influences of relative humidity. This is a **priority 1** instrument need, important since even the sign of the aerosol forcing depends on it.

- **Aerosol size distributions**. Knowledge of particle size distribution is essential to describe aerosol direct and indirect radiative forcing. Improved techniques are needed for determining the size distribution of ambient aerosols, including the influence of relative humidity. Current techniques are often ambiguous because of the assumption that particles are spherical. In addition optical techniques that are most often used in the 0.5-10 μm size range have inherent problems. Sizing techniques that are not based on optical properties need to be developed for particles in this size range. These techniques need to be tied to simultaneous measurements of properties such as mass, area (extinction) and number, so distributions can be reliably integrated. This is a **priority 2** instrumentation need.

III. RELATIONSHIP OF ASP TO OTHER PROGRAMS

The DOE Atmospheric Radiation Measurement Program supports research on quantifying the effect of aerosols on the radiation field, by investigating both the direct role of aerosols on radiative transfer and the indirect role on cloud properties. Specifically ARM research relates observations of radiative fluxes and radiances to the atmospheric composition and uses these relations to develop and test parameterizations to accurately predict the atmospheric radiative properties. In contrast, the ASP will support aerosol research with emphasis on aerosol processes and resulting properties that would influence the radiation fields. ASP scientists will be encouraged to utilize pertinent ARM data and to participate in field campaigns associated with ARM sites, both fixed and mobile. It is anticipated that the ARM mobile facility, currently under development, will at times be deployed in areas of interest specifically to ARM, at times in areas of interest specifically to ASP, and at times in areas of interest to both programs. It is anticipated that a joint ARM-ASP working group will be formed and collaborations between the two programs will be encouraged. Information about ARM can be found at http://www.arm.gov/

The DOE Climate Change Prediction Program (CCPP). The climate modeling community, as supported by DOE through the CCPP, is a major client for the research to be conducted in ASP. It is thus essential that ASP research be tailored to the needs of the climate modeling community and that the program provide specific, measurable, and meaningful deliverables. It is anticipated that a joint ASP-CCPP working group will be formed Information about CCPP can be found at http://www.science.doe.gov/ober/CCRD/model.html

NARSTO. Given the close linkages between aerosols, air quality, and climate change, ASP-funded scientists will be encouraged to participate in selected NARSTO field studies relevant to aerosol forcing. Information about NARSTO can be found at http://www.cgenv.com/Narsto/

NIGEC. Close coordination is anticipated between ASP and relevant research supported by NIGEC (National Institute for Global Environmental Change). NIGEC principal investigators will be invited to relevant ASP Science Team and working group meetings and encouraged to form collaborations with ASP scientists. Information about NIGEC can be found at http://www.science.doe.gov/ober/CCRD/nigec.html

Small Business Innovative Research (SBIR). The DOE Office of Science provides SBIR funding for instrument proposals. Information about SBIR can be found at http://sbir.er.doe.gov/sbir/ Eligible applicants are encouraged to apply under the FY 2005 SBIR/STTR Program Solicitations.

IV. SPECIAL CAPABILITIES OF DOE LABORATORIES

The DOE National Laboratories provide exceptional and unique capabilities that can contribute to the furtherance of the ASP objectives. ASP-funded scientists are encouraged to take advantage of these capabilities wherever practicable. ASP-funded scientists are also encouraged to form collaborations with DOE lab scientists and vice versa.

Following are among the relevant DOE laboratory capabilities. These and additional capabilities will be featured on the ASP website.

Research Aircraft Facility (RAF). It is anticipated that ASP will provide a small partially-instrumented manned aircraft (e.g., G-1) and/or instrumented unmanned aircraft in support of field studies. Other agencies may also provide instruments and aircraft platforms in collaboration with ASP. ASP scientists will be encouraged to deploy airborne and/or ground-based instrumentation and take measurements in conjunction with ASP and interagency field studies involving research aircraft. ASP field studies will be developed and coordinated with other federal agencies. Information on RAF capabilities can be found at http://www.pnl.gov/atmos_sciences/as_g1_2.html

The **Atmospheric Radiation Measurement** (ARM) Program is a multi-laboratory, interagency program that was created in 1989 with funding from the U.S. Department of Energy (DOE). The ARM Program is part of DOE's effort to resolve scientific uncertainties about global climate change with a specific focus on improving the performance of general circulation models (GCMs) used for climate research and prediction. These improved models will help scientists better understand the influences of human activities on the earth's climate. In pursuit of its goal, the ARM Program establishes and operates field research sites, called Cloud and Radiation Testbeds (CARTs), in several climatically significant locations. Scientists collect and analyze data obtained over extended periods of time from large arrays of instruments to study the effects and interactions of sunlight, radiant energy, and clouds on temperatures, weather, and climate. For additional information about ARM see http://www.arm.gov/docs/about.html

The Environmental Molecular Science Laboratory (EMSL) is a national user facility operated for DOE by Pacific Northwest National Laboratory. It offers a number of specialized instruments and capabilities (including high performance computing capabilities) for conducting field, laboratory and theoretical research on aerosol properties and processes. Proposers are encouraged to make use of these facilities as appropriate. Information on EMSL capabilities and user access can be found at http://www.emsl.pnl.gov/

The National Energy Research Scientific Computing Center (NERSC) at Lawrence Berkeley National Laboratory is one of the nation's most powerful unclassified computing resources and is a world leader in accelerating scientific discovery through computation. Funded by the Department of Energy's Office of Science (Mathematical, Information and Computational Sciences Division), NERSC currently serves about 2000 researchers at national laboratories, universities, and industries across the country. NERSC's mission is to accelerate the pace of scientific discovery in the DOE Office of Science community by providing high-performance computing, information, and communications services. NERSC is the principal provider of high performance computing services to Office of Science programs, including Environmental Research, and Advanced Scientific Computing Research. Information on NERSC can be found at http://www.nersc.gov/aboutnersc/intro.html.

The National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory in New York, is a national user research facility funded by the DOE's Office of Basic Energy Science. The NSLS operates two electron storage rings: an X-Ray ring and a Vacuum UltraViolet (VUV) ring which provide intense light spanning the electromagnetic spectrum from the infrared through x-rays. Each year over 2500 scientists from universities, industries and government labs perform research at the NSLS. Information on NSLS can be found at http://www.nsls.bnl.gov/

The Advanced Light Source (ALS), at Lawrence Berkeley National Laboratory, is a national user facility that generates intense light for scientific and technological research. As one of the world's brightest sources of ultraviolet and soft x-ray beams--and the world's first third-generation synchrotron light source in its energy range--the ALS makes previously impossible studies possible. The facility welcomes researchers from universities, industries, and government laboratories around the world. It is funded by the U.S. Department of Energy's Office of Basic Energy Sciences. Information on ALS can be found at http://www-als.lbl.gov/als/index.html

The **Advanced Photon Source** (APS) at Argonne National Laboratory is a national synchrotron-radiation light source research facility funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences. Using high-brilliance x-ray beams from the APS, members of the international synchrotron-radiation research community conduct forefront basic and applied research in the fields of material science; biological science; physics; chemistry; environmental, geophysical, and planetary science. Information on APS can be found at http://www.aps.anl.gov

CONTACTS FOR ADDITIONAL INFORMATION

Programmatic questions may be addressed to the Program Director, while questions pertaining to scientific scope may be addressed to either the Program Director or the Chief Scientist. We will try to provide answers in future FAQ's so that they are available to all interested parties. For this reason email is probably the best way to submit questions.

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